Magnetism and Atomic Structure.—I.

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The present communication .aims at an interpretation of the electron structure of matter which is compatible with the results of recent magnetic investigations.   While acknowledging in full the success of the Rutherford-Bohr-Sommerfeld theory, in so far as it applies to radiation problems, it must at the same time be admitted that this success is confined to the simplest types of atoms, the hydrogen atom and the positively charged helium atom, each with a single electron, such atoms being very probably in an abnormal state as compared with that which prevails in unexcited matter, and upon which the magnetic determinations have been conducted.   As a consequence, we are led to somewhat different interpretations of atomic structure, according to which viewpoint is adopted; but a fuller recognition of the possible differences of atomic structure between radiating and non-radiating matter may, in the near future, enable us to bridge the gap which at the moment unmistakably divides them.

This paper* is a continuation of previous memoirs† on "The Influence of Molecular Constitution and Temperature on Magnetic Susceptibility" ('Phil. Trans.,' A, vol. 214, p. 109, 1914, Parts I and II; vol. 215, p. 79, 1915, Part III; vol. 220, p. 247, 1920, Part IV; 'Proc. Roy. Soc.,' A, vol. 95, p. 58, 1918).   In Part III, p. 92, and Part IV, pp. 27 0–276, I have called attention to the classical experiments of Tyndall‡ on the deportment of crystalline bodies in the magnetic field.   From observations on about 100 different crystals suspended in a uniform magnetic field, Tyndall showed that, in general, "if the arrangement of the component particles of any body be such as to present different degrees of proximity in different directions, then the line of closest proximity, other circumstances being equal, will be that chosen by the respective forces for the exhibition of their greatest energy.   If the mass be magnetic, this line will stand axial; if diamagnetic, equatorial."   Tyndall propounded the "theory of reciprocal induction" to account for the augmented diamagnetic or paramagnetic property in the direction of closest packing of the particles.   At that time

* Abstract read before the British Association, Cardiff Meeting, August 27, 1920.

† A *résumé* of these papers is given in 'Science Progress,' No. 56, pp. 588–601 (March, 1920).

‡ 'On Diamagnetism and Magnecrystallic Action,' p. 23 (1870).

(1870) the diamagnetic forces were known to be so minute that the theory of reciprocal induction, as applied to minute diamagnets, appeared incredible, and, as a correspondence between Lord Kelvin and Tyndall shows,* the former expressed emphatically his opinion that this theory was quite incapable of accounting quantitatively for the effects observed. Regarding a diamagnetic molecule as a simple diamagnet, the mutual actions between such systems would certainly be inadequate. It has, however, been shown (Part II, p. 143; Part III, pp. 83–92) that a diamagnetic molecule is not a simple but a complex diamagnet, formed of at least two components, each of which is strongly magnetic locally. These components are the rotating electrons, whose equivalent magnetic moments are

$$\mathrm{M} = \frac{\epsilon \mathrm{S}}{\tau},$$

where ϵ is the electronic charge, S/τ the aërial velocity with which the orbit of area S is described in time τ. Since in different types of atoms the electrons are comparable in number, and revolve in orbits for which S and τ are of the same order, the local magnetic fields will be of comparable magnitude whether the atoms be diamagnetic or paramagnetic (see figs. 1a and 1b). Such strong local forces, which are of the order 10^7 gauss,† will account quantitatively, on the theory of reciprocal induction, for the directive effects observed by Tyndall.
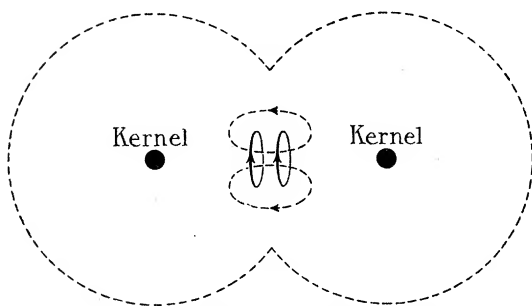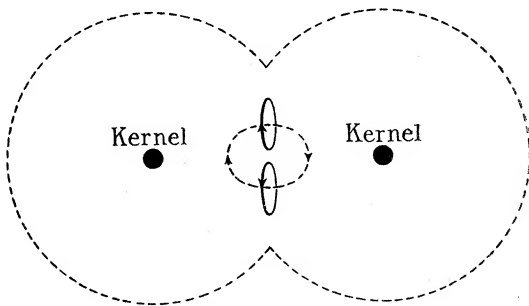

FIG. 1a.—Paramagnetic coupling. FIG. 1b.—Diamagnetic coupling.
The black dots indicate the diffuse atomic kernels, consisting of positive charges and a spacial distribution of electrons, which are rotating in small circles at varying distances from the nuclei.

In a crystal, the direction of closest packing of the molecules is parallel to the principal cleavage, and, if the remaining cleavages are comparatively insignificant, this direction sets equatorially with respect to the magnetic field if the crystal is diamagnetic, and axially if it is paramagnetic. This is

* *Loc. cit.*, Various Letters, pp. 221–234.
† 'Phil. Trans.,' A, vol. 215, pp. 84–87 (1915).

true, within limits, even when the axial dimension of the crystal is greatest in the former case, and the equatorial dimension greatest in the latter case; the diamagnetic crystal thus behaving as regards its outward form like a paramagnetic body, and the paramagnetic crystal like a diamagnetic body. This difference of deportment is shown in fig. 2. In general, other
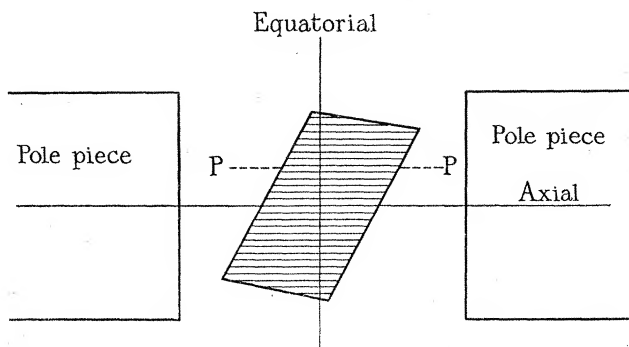

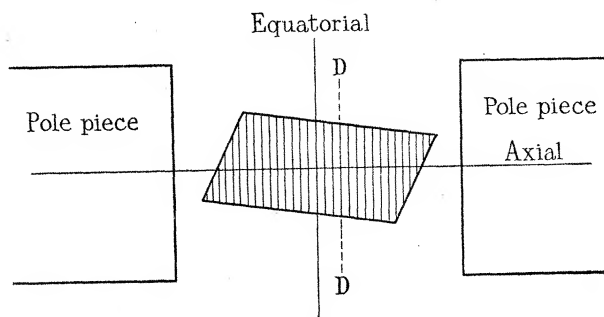Fig. 2.—Paramagnetic crystal. Direction of principal plane of cleavage, PP, is axial.

Fig. 2.—Diamagnetic crystal. Direction of principal plane of cleavage, DD, is equatorial.

cleavages than the principal one must be taken into account, and on this Tyndall remarks: "When a crystal cleaves symmetrically in several planes, all parallel to the same straight line, and, at the same time, in a direction perpendicular to this line, then the latter cleavage, if it be more eminent than the former, may be expected to predominate; but, when the cleavages are oblique to each other, the united action of several minor cleavages may be such as to overcome the principal one, or so to modify it that its action is not at all the same as that of a cleavage of the same value unintersected by others."

Recently, I have had an opportunity of extending some of Tyndall's experiments to the following organic compounds: naphthalene,* anthracene,

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sodium-ammonium racemate, tartaric and citric acids. The effects observed were in agreement with the rules given by Tyndall for other crystalline substances, and it will be sufficient to describe here those obtained with naphthalene, which possesses a very pronounced cleavage. This substance, which is diamagnetic, crystallises in thin plates having monoclinic symmetry. Three piles of these plates were constructed by cementing together selected crystals with Canada balsam (diamagnetic). The numbers of crystalline plates in the built up specimens were (a) two, (b) forty, (c) about eighty ; in (b) and (c), the orientations of the plates, about a direction perpendicular to the principal cleavage, being arranged at random. Each specimen was suspended by a 50-cm. length of unspun silk in a uniform magnetic field of approximately 10,000 gauss. The deportment of each specimen is shown in plan in fig. 3.

The principal plane of cleavage invariably sets itself equatorial, even though in cases (b) and (c) the ratios

FIG. 3.—Deportment of naphthalene crystals in magnetic field. (Plan.)

of the axial to the equatorial dimensions were 1 : 1 and 1·75 : 1 respectively; the specimen (c) thus behaving like a paramagnetic body, in so far as its outward form is concerned, setting its longer dimension from pole to pole. The specimens were diamagnetic, and were definitely repelled from one pole in a non-uniform field. The deportment was in each case decisive, the complete period of oscillation being 10 seconds for specimen (c) with the field off, and 1·5 seconds only with the field on. The specimens could readily be caught with either end opposite the north pole of the electromagnet.

The crystalline structure is evidently so constituted that the diamagnetic effect is a maximum parallel to the principal cleavage, and in this direction also the cohesive force is a maximum. I have, in a previous paper,* attributed this cohesive force to the mutual induction between a pair of electron orbits, for it appears that, in organic compounds where there is no
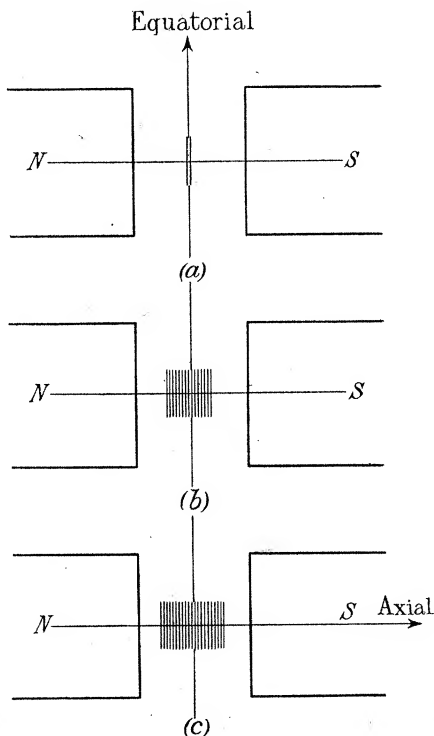
* 'Phil. Trans.,' A, vol. 215, p. 83 (1915).

ionisation, and in elements composed of identical particles, it is only by such means that attraction can be secured at all (unless one assumes the existence of some hypothetical constitutive force whose origin is entirely unknown and consequently does not help us).

The coupling is of the nature shown in figs. 1a and 1b, the latter type of coupling being necessary in the case of the hydrogen molecule, containing only two electrons, in order to preserve its diamagnetism. A suggested model of the hydrogen molecule* is illustrated in fig. 4. The pair of electrons,
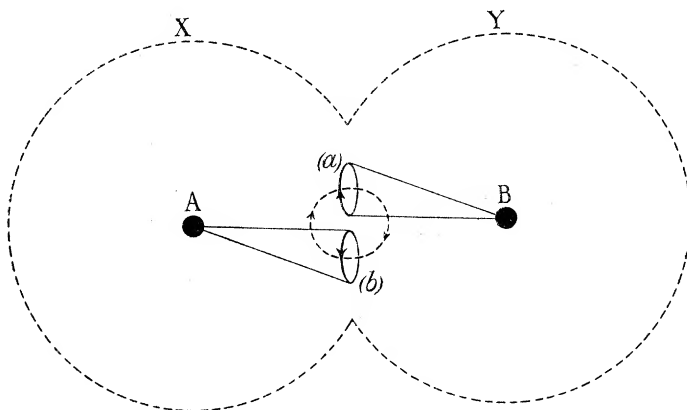

FIG. 4.—Model of hydrogen molecule.

a and b, held in common by two exactly similar atoms, X and Y, constitute an astatic system, each electron revolving in a small circular orbit (or
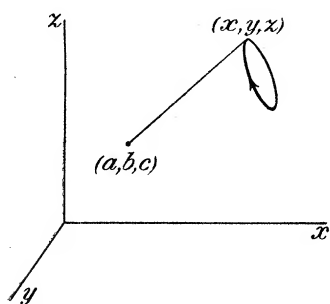

FIG. 5.

possibly an ellipse) and being completely, or more probably almost completely, bound to its own nucleus by a tube of force.† The equilibrium of the electron orbits is one in which a balance is secured between the residual electrostatic repulsive forces and the magnetic attractive forces.‡

As regards the deportment of an unsymmetrical diamagnetic atom in a magnetic field, let x, y, z be the co-ordinates of an electron,

* A. E. Oxley, 'Nature,' vol. 105, pp. 327–328 (May 13, 1920).

† It will be observed that each hydrogen atom will, on this view, be paramagnetic. The ferromagnetism of manganese (Sir Robert Hadfield, Chéneveau, and Géneau, 'Roy. Soc. Proc.,' A, vol. 94, p. 65 (1917)), fused in an atmosphere of hydrogen, may possibly be attributed to occluded hydrogen in the atomic state. It is an interesting fact that hydrogen is readily occluded by other highly magnetic elements.

‡ Cf. 'Phil. Trans.,' A, vol. 220, p. 289 (1920).

a, b, c those of the nucleus or centre of gravity of the atom (fig. 5). Writing

$$(\xi, \eta, \zeta) = (x-a, y-b, z-c),$$

then the induced diamagnetic moment, ΔM_z, due to the application of an external field H_z parallel to O_z is

$$\Delta M_z = -\frac{\epsilon^2 \cdot H_z \cdot A}{4m},* \tag{1}$$

where

$$A/2 = \Sigma\xi^2 = \Sigma\eta^2 = \Sigma\zeta^2$$

and ϵ/m is the ratio of the charge to the mass of an electron. This holds for any type of orbit. If the orbit is in a plane parallel to H_z, ΔM_2 is zero; for any other orientation of the orbit, ΔM_z is negative, whatever the direction of revolution of the electrons, because (1) does not depend on the angular velocity. In the case of a diamagnetic coupling system like that shown in fig. 1b, the pair of orbits would tend to set with their common plane parallel to the magnetic field. In a crystal, each molecule, except for small thermal oscillations, presents a constant orientation to its neighbours, and the cohesive or coupling forces will in general be stronger in some directions than in others. Thus, in the case of the diamagnetic naphthalene crystals, providing the electron coupling is strongest in a direction parallel to the principal plane of cleavage, due to a closer interaction between molecules which lie in a line parallel to this plane, we should expect the planes of these coupling orbits to set axially, *i.e.*, the plane of cleavage should set equatorially, as it actually does. Any departure from this setting would produce an increased diamagnetic effect, causing the crystal to return to the equatorial position, which is therefore stable. In general there will be structural forces in other directions, but where the principal cleavage is so pronounced, as it is in naphthalene, the deportment of the crystal in the magnetic field will isolate the principal cleavage.

On the other hand, in paramagnetic crystals, where the principal cleavage is determined by a paramagnetic pair of coupling orbits, as shown in fig. 1a this cleavage will set axially with respect to the magnetic field. Any displacement from this position results in an increase of potential energy and therefore this position of equilibrium is also stable.

In other words, both in diamagnetic and paramagnetic media, the planes of the electron orbits have a greater aggregate projection perpendicular to the principal cleavage than parallel to it. As a consequence, the existence of several cleavages, even though they be minor ones, demands a spacial distribution of electron orbits about the atomic nuclei.

* 'Phil. Trans.,' A, vol. 214, p. 137 (1914); and references to Langevin's paper there given.

It appears from these considerations that, except perhaps for crystals of compounds of the simple cubic form, it is possible to isolate the fundamental unit of the crystal structure by observing the equilibrium positions of a crystal when suspended in dffierent ways in a magnetic field. These equilibrium positions are consistent with the disposition of the planes of cleavage, *i.e.*, with the closer or more open packing of the molecules in certain directions. In such cases *the fundamental unit of the crystal structure is the molecule.**

Recently, the existence of special relationships between pairs of electrons has been supplied by X-ray work,† but it seems doubtful if definite evidence as to the nature of the electron coupling can be safely drawn from this source until the effects of thermal oscillations on the intensity of the diffraction effects have been investigated.

In a simple cubic crystal like rock-salt, the X-ray analysis indicates that all the atoms of one kind are equally influenced by their symmetrically disposed neighbours.‡ It is interesting to note that rock-salt shows no appreciable structural deportment in the magnetic field, the evidence of three equally important cleavages at right angles to one another satisfactorily accounting for this.

The views on atomic structure which have been given above are consistent with the theory of the cubical atom first suggested by G. N. Lewis§ and

* *Cf.* 'Phil. Trans.,' A, vol. 220, p. 274 (1920); 'Science Progress,' No. 56, p. 596 (1920).

† Debye and Scherrer ('Phys. Zeits.,' vol. 18, p. 291 (1917)) have examined the structure of graphite, and from their results, W. L. Bragg ('Phil. Mag.,' vol. 40, p. 185 (1920)), has suggested that certain of the atoms are sharing electrons. In a later paper, Debye and Scherrer ('Phys. Zeits.,' vol. 19, p. 47 (1918)) have considered the structure of diamond and consider that such pairs of coupling orbits are inconsistent with the experimental work of the Braggs. This conclusion is criticised by D. Coster ('K. Akad. Wetensch. Amsterdam,' vol. 22, p. 1 (1919)), who points out that if the different orientations of the planes of the coupling orbits are taken into account, it is impossible in the present experimental stage to draw any definite conclusions from the X-ray diffraction experiments as to the nature or existence of the coupling electrons. James and Tunstall ('Phil. Mag.,' vol. 40, p. 239 (1920)) show that in antimony, the planes of atoms are disposed in pairs. Taking the close pair, and one atom in each, the nearest distance between atomic centres is 2·87 Å.U. Taking the wide pair, the corresponding nearest distance is 3·37 Å.U. They remark that the very good cleavage of antimony is parallel to these pairs of planes. Antimony possesses other cleavages, and it is possible that similar, though smaller, differences of atomic distances exist for these minor cleavages, and, if so, we may expect direct evidence of the isolation of the molecule in crystalline antimony.

‡ *Cf.*, however, Thirring, 'Phys. Zeits.,' vol. 21, p. 281 (1920), in connection with the crystal lattices of rock salt and sylvine. Thirring introduces magnetic forces to account for the hemihedrism of sylvine.

§ 'J. Amer. Chem. Soc.,' vol. 38, p. 72 (1916).

afterwards considerably extended by Irving Langmuir.* This theory presents certain analogies to Bohr's theory, but differs from the latter in respect of a spacial distribution of the electrons about the nucleus. The electrons must, however, be rotating ;† possibly in small circles or ellipses at various distances from the nucleus, in order to account for permanent magnetism and optical activity,‡ as suggested in previous work. Alternatively, the electron itself may be a complex unit endowed with specific magnetic as well as electrostatic properties ; this would be equivalent magnetically to a revolving electron and might in some respects be more satisfactory. In either case, during the process of radiation, the electron passes discontinuously through a series of states; the positions of equilibrium, viz., those defining the normal atom, being spherical or ellipsoidal surfaces on each of which the electrons form a space pattern. This would be the three-dimensional interpretation of the mechanism of radiation suggested by Bohr's theory.

According to the Lewis-Langmuir theory, the most stable electron grouping, in elements and in compounds of the electro-negative elements, is a pair of electrons. The next most stable arrangement is the octet, *i.e.*, eight electrons arranged at the corners of a cube. This cube is often very much distorted, and " when a pair of electrons is held in common between two atoms, the chemical evidence indicates that the pair acts as a unit." Such a pair constitutes the single valency bond.§ Lewis's arguments for the close approach of the electrons in the pair, which we have attributed to magnetic induction, are based upon a discussion of :—(1) The origin of the triple bond, due to the pairing of the eight electrons of the octet so as to form a tetrahedron ; (2) The interpretation of the Baeyer strain theory ; (3) The explanation of the free mobility about a single bond which must always be assumed in stereo-chemistry.

The cubical atom theory gives us a much clearer picture of the nature of diamagnetism and paramagnetism than the Bohr-Sommerfeld theory, with its large unbalanced electron orbits, can give at present, and it appears that a similar remark applies to the interpretation of crystal structure.‖ Born and Landé¶ have studied the compressibilities of the alkali halides, on Bohr's assumptions, and conclude that the electron orbits do not lie in a plane, but

* *Loc. cit.*, vol. 41, p. 868 (1919).

† ' Phil. Trans.,' A, vol. 220, pp. 274, 289 (1920) ; A, vol. 215, p. 87 (1915). ' Nature,' vol. 105, p. 105 (1920).

‡ W. E. Garner, ' Nature,' vol. 104, p. 661 (1920) ; vol. 105, p. 171 (1920).

§ *Cf.* ' Phil. Trans.,' A, vol. 220, pp. 277–280 (1920).

‖ W. L. Bragg, ' Phil. Mag.,' vol. 40, p. 169 (1920).

¶ ' Verh. Deutsch. Phys. Gesell.,' vol. 20, pp. 210, 230 (1918).

are arranged in space with cubic symmetry.* Sommerfeld also expresses the view that such a conception may eventually enable us to overcome certain outstanding difficulties connected with spectra.

Magnecrystallic action, as we have seen, demands a spacial distribution of electron orbits. All diamagnetic substances (and these form by far the larger proportion of substances known to us) must be internally self-compensated. This may be secured by a symmetrical distribution of electron orbits.

Paramagnetic molecules, in general, have unbalanced magnetic moments. This does not imply that on crystallisation ferro-magnetism would result. On the lines of the cubic atom theory, it is easy to extend Ewing's model to atom groups of eight elementary magnets, each of which has a definite resultant moment, and to arrange systems of such groups so that the crystalline aggregate possesses a zero magnetic moment in the absence of an external field. When a field is applied, a re-distribution of the resultant magnetic axes of the groups will take place, accompanied by the appearance of paramagnetism, which may be different along specialised directions and which will depend upon the temperature.

Ferro-magnetism is a limiting case, depending on a crystalline molecular grouping, which is unstable for fields above a certain strength, and in which, when this field strength has been reached, saturation and permanent magnetisation are produced, either by a rotation of certain molecules as a whole, or by a rotation of the planes of individual electron orbits in those molecules.

On the views which have been expressed above, the rigidity of crystalline media is due to the highly localised nature of the small magnetic coupling elements. A relative rotation of the two coupling circuits through an angle comparable with 45° would so weaken the coupling, due to the transverse separation of the minute magnetic doublets, that the crystalline cohesion would disappear. Consequently, a sufficient rise of temperature to cause such oscillations would result in fusion, the isotropic liquid produced having no rigidity. Such oscillations would have very little effect on the electro-static stress between the molecules, since the electrostatic doublets are comparable in length with atomic dimensions. It is suggested that the Laplace intrinsic pressure is due to this electrostatic stress. Crystallisation is accompanied by a directed magnetic stress, which, superimposed upon the Laplace pressure, gives rise to a crystalline symmetry characteristic of the molecular configuration. The rigidity of isotropic gels, as pointed out in 'Phil. Trans. Roy. Soc.,' A, vol. 215, p. 81, 1915, is of a different nature, and

* 'Atombau u. Spectrallinien,' 1919, p. 368.

is due to an interlocking of molecules, whose axes are arranged at random, when the thermal agitation is sufficiently and quickly reduced.

Conclusions.

The close relationship which exists between the equilibrium positions of diamagnetic and paramagnetic crystalline bodies, suspended in a magnetic field, and the directions of the cleavage planes can be satisfactorily interpreted in terms of a spacial distribution of electron orbits round the nucleus. The orbits may have any shape, but are probably small in comparison with atomic dimensions. In un-ionised media there is evidence that the coupling force between the units of the crystalline structure is that of magnetic induction; the mechanical stress accompanying it being balanced by the stress due to the distortion of the internal electrostatic configuration of the units. The coupling systems, which account for these attractions, are pairs of electron orbits. In this respect the present theory is closely allied to the cubical atom theory of Lewis and Langmuir, in which the most stable electron arrangement is that of a pair.

Tyndall's discovery relating to the characteristic deportment of diamagnetic and paramagnetic bodies in the magnetic field has been confirmed for a few organic compounds, and the interpretation of the results on the electron theory implies that, both in diamagnetic and paramagnetic crystals, the planes of the electron orbits have a greater aggregate projection perpendicular to the principal cleavage than parallel to it. Thus the existence of multiple cleavages demands a spacial distribution of electron orbits. Moreover, as the molecules are closest packed in a direction parallel to the principal cleavage, the isolation of all the cleavages by suitably suspending the crystal in a magnetic field really isolates the unit of the crystal structure. This unit is the chemical molecule. It is interesting to note that crystals of the simple cubic form, for which X-ray analysis indicates an ionised atomic structure, rather than a molecular structure, show no appreciable structural deportment in the magnetic field.

It is probable that the atoms of hydrogen and helium, the structure of which has been so successfully attacked from the radiation standpoint by Bohr and Sommerfeld, are, when radiating, in a state very different from the unexcited matter upon which magnetic observations have been made. In magnetic work we are concerned with matter in what we may call the "normal state," each atom of which contains at least one electron within the active range of another electron in an adjacent atom, and the mutual interaction of such systems must be taken into account. Some factor seems to have been neglected in Bohr's theory, which, though of small importance

when the atoms contain each only one electron and are independent of each other, yet is of primary importance for matter in its normal state, and without which it is impossible to explain either the magnetic properties of crystals or crystal structure.   The attraction between exactly similar atoms, the directed valencies required by stereo-chemistry, and the directive forces within crystals are explicable only by a due recognition of the magnetic forcive in material media.

It is probable that the Laplace intrinsic pressure in liquids and in isotropic gels is due to electrostatic doublets of atomic dimensions.   When crystallisation sets in, superimposed upon the electrostatic stresses, are the directional stresses due to the magnetic doublets which, for a given molecule, determine a characteristic crystalline symmetry, and, owing to their highly localised nature compared with the electrostatic doublets, determine the rigidity of the crystal.   Such highly localised magnetic doublets suggest either that the conventional electron is moving in an orbit very small compared with atomic dimensions, or else that the electron itself is a complex unit endowed with specific magnetic as well as electrostatic properties, *i.e.*, the electron is also a magneton (*supra*, p. 271).

On the Measurement of Low Magnetic Susceptibility by an Instrument of New Type.

By ERNEST WILSON, M.Inst.C.E., M.Inst.E.E.

It is well known in instruments of the Curie type that when the magnetising force impressed upon the substance is varied and the mechanical force is balanced against that of the torsion in a suspending fibre, the deflections follow the square law and the range through which the magnetic force can be varied is restricted.   In fact, it is difficult with the Curie balance to measure the susceptibility of a given specimen outside a narrow range of magnetic force of about one to five.   One object in the design of the present instrument was to be able to measure susceptibility with a given specimen through a wide range of magnetic force.   Another object was to make the instrument portable in the sense that no spot of light and scale are required, and yet be able to measure susceptibility through a wide range.   In all the tests so far made the instrument was placed on an ordinary table.